Testing quantum correlations versus single-particle properties within Leggett's model and beyond

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Quantum theory predicts and experiments confirm that nature can produce correlations between distant events that are nonlocal in the sense of violating a Bell inequality [1]. Nevertheless, Bell's strong sentence *Correlations cry out for explanations* **remains relevant. The maturing of quantum information science and the discovery of the power of nonlocal correlations, e.g. for cryptographic key distribution beyond the standard Quantum Key Distribution schemes [2, 3, 4], strengthen Bell's wish and make it even more timely.**

In 2003, Leggett proposed an alternative model for nonlocal correlations [5], that he proved to be incompatible with quantum predictions. We present here a new approach to this model, along with new inequalities for testing it. Remarkably these inequalities can be derived in a very simple way, assuming only the non-negativity of probability distributions; they are also stronger than previously published Leggett-type inequalities [5, 6, 7, 8]. The simplest of these inequalities is experimentally violated. Then we go beyond Leggett's model, and show that one cannot ascribe even partially defined individual properties to the components of a maximally entangled pair.

Formally, a correlation is a conditional probability distribution $P(\alpha, \beta | \vec{a}, \vec{b})$, where α, β are the outcomes observed by two partners, Alice and Bob, when they perform measurements labeled by $\vec{a}$ and $\vec{b}$, respectively. On the abstract level, $\vec{a}$ and $\vec{b}$ are merely inputs, freely and independently chosen by Alice and Bob. On a more physical level, Alice and Bob hold two subsystems of a quantum state; in the simple case of qubits, the inputs are naturally characterized by vectors on the Poincaré sphere, hence the notation $\vec{a}, \vec{b}$.

How should one understand nonlocal correlations, in particular those corresponding to entangled quantum states? A natural approach consists in decomposing $P(\alpha, \beta | \vec{a}, \vec{b})$ into a statistical mixture of hopefully simpler correlations:

$$P(\alpha, \beta | \vec{a}, \vec{b}) = \int \mathrm{d}\lambda \rho(\lambda) \, P_\lambda(\alpha, \beta | \vec{a}, \vec{b}) . \qquad (1)$$

Bell's locality assumption is $P_\lambda(\alpha, \beta | \vec{a}, \vec{b}) = P_\lambda(\alpha | \vec{a}) P_\lambda(\beta | \vec{b})$; admittedly the simplest choice, but an inadequate one as it turns out: quantum correlations violate Bell's locality [1]. Setting out to explore other choices, it is natural to require first that the P_λ's fulfill the so-called *no-signaling condition*, i.e., that none of the correlations P_λ results from a communication between Alice and Bob. This can be guaranteed by ensuring space-like separation between Alice and Bob. Non-signaling correlations happen without any time-ordering: there is not a first event, let's say at Alice's side, that causes the second event via some *spooky action at a distance*. One may phrase it differently: non-signaling correlations happen from outside space-time, in the sense that there is no story in space-time that tells us how they happen. This is the case in orthodox quantum physics, or in some illuminating toy models like the nonlocal box of Popescu and Rohrlich (PR-box) [9]. Mathematically, the no-signaling condition reads $P_\lambda(\alpha | \vec{a}, \vec{b}) = P_\lambda(\alpha | \vec{a})$ and $P_\lambda(\beta | \vec{a}, \vec{b}) = P_\lambda(\beta | \vec{b})$: the local statistics of Alice are not influenced by Bob's choice of measurement, and reciprocally.

In 2003, Leggett proposed another model of the form (1), which can also be experimentally tested against quantum predictions [5]. This model was recently brought into focus by the work of Gröblacher *et al.* [6]. The basic assumption of Leggett's model is that locally everything happens as if each single quantum system would always be in a pure state. We shall be concerned here with the case of binary outcomes $\alpha, \beta = \pm 1$, though generalizations are possible. In this case, the supplementary variables λ in Leggett's model describe pure product states of two qubits, denoted by normalized vectors $\vec{u}, \vec{v}$ on the Poincaré sphere:

$$\lambda = |\vec{u}\rangle \otimes |\vec{v}\rangle , \qquad (2)$$

and the local expectation values have the usual form as predicted by quantum physics:

$$\langle \alpha \rangle_\lambda = \langle \vec{u} | \vec{a} \cdot \vec{\sigma} | \vec{u} \rangle = \vec{u} \cdot \vec{a} , \qquad (3)$$
$$\langle \beta \rangle_\lambda = \langle \vec{v} | \vec{b} \cdot \vec{\sigma} | \vec{v} \rangle = \vec{v} \cdot \vec{b} . \qquad (4)$$

If the qubits are encoded in the polarization of photons, as in Leggett's initial idea, then the assumption is that

each photon should locally behave as if it were perfectly polarized (in the directions $\vec{u}$ and $\vec{v}$), and the local observations, conditioned on each λ, should fulfill Malus' law. It is worth emphasizing that Leggett's assumption concerns exclusively the local marginals $\langle\alpha\rangle_\lambda$ and $\langle\beta\rangle_\lambda$ of the probability distributions P_λ, while nothing is specified about the correlation coefficients $\langle\alpha\beta\rangle_\lambda$. Leggett's model can thus still be nonlocal, and can in general violate a Bell inequality.

Both in the original paper [5] and in [6], the model was presented by implicitly assuming a time-ordering of the events. Any model based on such an assumption had already been falsified by the so-called before-before experiment [10, 11], as Suarez emphatically stressed [12]. However, assumptions (3–4) clearly define non-signaling correlations, and Leggett's model can be defined without any reference to time-ordering. As a consequence, its study does add something to our understanding of quantum non-locality. But what exactly? In what are such P_λ's "simpler" than the usual quantum correlations? To answer these questions, we recall that, in quantum theory, the singlet state is such that the properties of the pair are sharply defined (the state is pure), but the properties of the individual qubits are not. In this perspective, Leggett's model is an attempt of keeping the correlations while reintroducing sharp properties at the individual level as well.

Leggett's model cannot reproduce the correlations of the singlet state. Experimental falsifications have already been reported, first under additional assumptions [6], then more directly [7, 8]. These works relied on the violation of so-called *Leggett-type inequalities*. Analog to Bell's inequalities, these criteria say that, under Leggett's assumptions (3) and (4), a measurable quantity L should satisfy $L \leq L_{\max}$, while quantum theory predicts that $L > L_{\max}$ can be observed for suitable measurements. An important feature of Leggett-type inequalities is that, contrary to Bell's inequalities, the bound L_{max} is not a fixed number: instead, like the model itself, it depends on the quantum measurements that are performed. Consequently all experimental data aiming at disproving Leggett's model should present evidence that the settings used in the experiment have been properly adjusted.

All previously available derivations of Leggett-type inequalities were quite lengthy and failed to suggest possible improvements or generalizations. We have found a much more straightforward derivation (see the Methods section), simply based on the fact that each P_λ must be a valid probability distribution, so in particular $P_\lambda(\alpha, \beta|\vec{a}, \vec{b}) \geq 0$. Remarkably, this constraint of *non-negativity of probabilities*, weak as it may seem, is enough to induce an observable incompatibility between Leggett's model and quantum predictions. In our derivation, it also appears that the previously derived Leggett-type inequalities are sub-optimal; among the improved inequalities that our approach suggests, the simplest one
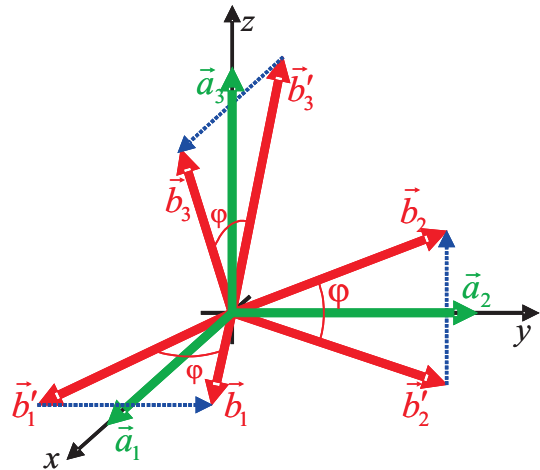

FIG. 1: **Alice's (green) and Bob's (red) measurement settings used to test inequality (5).** In order for inequality (5) to hold, Bob's settings must be such that the three pairs $(\vec{b}_i, \vec{b}'_i)$ form the same angle φ, and the three directions $\vec{e}_i$ of $\vec{b}_i - \vec{b}'_i$ (blue) are orthogonal. The best violation is obtained when Alice's settings $\vec{a}_i$ are chosen to be along the directions of $\vec{b}_i + \vec{b}'_i$.

reads

$$\frac{1}{3}\sum_{i=1}^{3}|C(\vec{a}_i, \vec{b}_i) + C(\vec{a}_i, \vec{b}'_i)|$$

$$\equiv L_3(\varphi) \;\leq\; 2 - \frac{2}{3}|\sin\frac{\varphi}{2}| \quad (5)$$

where $C(\vec{a}, \vec{b}) = \sum_{\alpha,\beta}\alpha\beta P(\alpha, \beta|\vec{a}, \vec{b})$ is the usual correlation coefficient. This inequality holds provided the three measurements on Alice's side and six on Bob's fulfill some relations; a possible set of measurements is given in Figure 1. For the singlet state, quantum mechanics predicts $C_{\Psi^-}(\vec{a}, \vec{b}) = -\vec{a}\cdot\vec{b}$. Thus, for the settings just defined, $L_3(\varphi)$ is

$$L_{\Psi^-}(\varphi) = 2|\cos\frac{\varphi}{2}| \;. \quad (6)$$

This expression violates inequality (5) for a large range of values φ.

In order to test the Leggett-type inequality (5) in an experiment, we prepared polarization-entangled photon pairs into single mode optical fibers in a close approximation to a singlet state, similarly as in [8]. In our setup (see Figure 2), we choose the settings of polarization measurements $\vec{a}_i, \vec{b}_i$ and $\vec{b}'_i$ for the individual photons by dialing in appropriate orientation angles $\gamma_{A,B}$ for two quarter wave plates ($\lambda/4$), and angles $\theta_{A,B}$ for two absorptive polarization filters (PF). Details about the experimental implementation can be found in Supplementary Information I. Through four consecutive coincidence measurements between photodetectors $D_{A,B}$ for all combinations
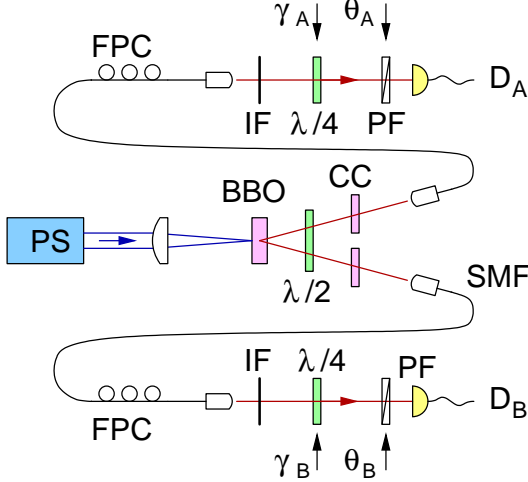
FIG. 2: **Experimental setup.** Polarization-entangled photon pairs are generated in a nonlinear optical crystal (BBO) and coupled into single mode optical fibers (SMF), similarly as in [8]. Polarization measurements in arbitrary settings for each photon are performed with polarization filters (PF) and quarter wave plates ($\lambda/4$), followed by single photon detectors ($D_{A,B}$). More details can be found in Supplementary Information I.
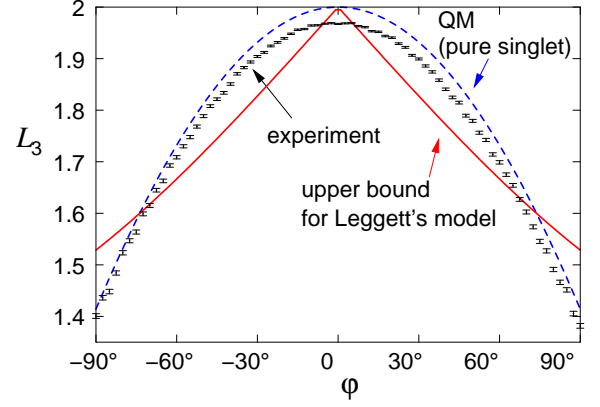

FIG. 3: **Violation of Leggett's model.** The experimental values for L_3 over a range of separation angles φ (points with error bars) violate the bound given by Leggett's model (solid line), and follow qualitatively the quantum mechanical (QM) prediction (dashed line). The error bars indicate ± 1 standard deviations of propagated Poissonian counting statistics assumed for photodetection events. The largest violations of inequality (5) are found for $\varphi = \pm 25°$ with 40.6 and 38.1 standard deviations, respectively.

of settings $\vec{a}, -\vec{a}$ and $\vec{b}, -\vec{b}$, we establish an experimental value for a correlation coefficient $C(\vec{a}, \vec{b})$.

The correlation coefficients necessary to compose values for $L_3(\varphi = \pm 30°)$ were obtained with an integration time of $T = 60$ s per point, leading to values of 1.9068 ± 0.0009 for $\varphi = -30°$ and 1.9005 ± 0.0010 for $\varphi = 30°$. This corresponds to a violation of the bound for $L_3(\varphi = -30°)$ and $L_3(\varphi = +30°)$ in Leggett's model by 83.7 and 74.5 standard deviations, respectively.

The asymmetry in the measured values of $L_3(\varphi)$ is an indication for experimental imperfections in the accuracy of the settings, such as a possible misalignment of one of the quarter wave plates with respect to the polarizing filters. To test this alignment, we collected values for L_3 over a larger range of φ with an integration time of $T = 15$ s per setting (Figure 3). The variation of L_3 with φ is compatible with the quantum mechanical prediction for a singlet state with residual colored noise and an orientation uncertainty of the quarter wave plate of $0.2°$.

The falsification of Leggett's model proves that it is impossible to reconstruct quantum correlations from hypothetical, more elementary correlations in which individual properties would be sharply defined. Let us argue that a much stronger statement holds, namely, that individual properties cannot even be partially defined.

We first consider the following straightforward generalization of Leggett's model: we allow the "local states" λ to be mixed states, e.g. photons with a degree of polarization η. So, we replace (3) and (4) by

$$\begin{aligned} \langle \alpha \rangle_\lambda &= \eta \, \vec{u} \cdot \vec{a}, \\ \langle \beta \rangle_\lambda &= \eta \, \vec{v} \cdot \vec{b}, \end{aligned} \quad \text{with } 0 < \eta \le 1. \quad (7)$$

The derivation of inequalities for this model follows exactly the same pattern as for usual Leggett-type inequalities. In particular, the generalized version of (5) is

$$L_3(\varphi) \le 2 - \frac{2}{3} \eta \, |\sin \frac{\varphi}{2}| \quad (8)$$

which, for angles φ small enough, is violated by $L_{\Psi^-}(\varphi)$ for any value of $\eta > 0$. Thus, as soon as the degree of purity of the "local states" is non-zero, this generalization of Leggett's model also fails to reproduce quantum mechanical predictions (see Supplementary Information II for a more complete analysis of this generalization).

Experimentally, one cannot expect to conduct a meaningful comparison between those two predictions down to $\eta = 0$, due to imperfections in the state preparation. From the measurement of Figure 3, however, we can claim experimental evidence of a violation for all $\eta \ge 0.56$, with a statistical significance of at least 3.65 standard deviations, thus putting a lower bound for this class of models (see Figure 4).

It is then natural to conjecture that no model of form (1), with non-signaling correlations P_λ, can perfectly reproduce the correlations of the singlet state $C_{\Psi^-}(\vec{a}, \vec{b}) = -\vec{a} \cdot \vec{b}$, unless

$$\langle \alpha \rangle_\lambda = \langle \beta \rangle_\lambda = 0 \quad (9)$$

for all measurements $\vec{a}$ and $\vec{b}$ (except perhaps for a few λ's in a zero-measure set). In the Methods section we prove this conjecture for models with discrete supplementary variables λ; continuity arguments should allow to extend the result to more general models. We thus have a necessary condition for a non-signaling model to be compatible
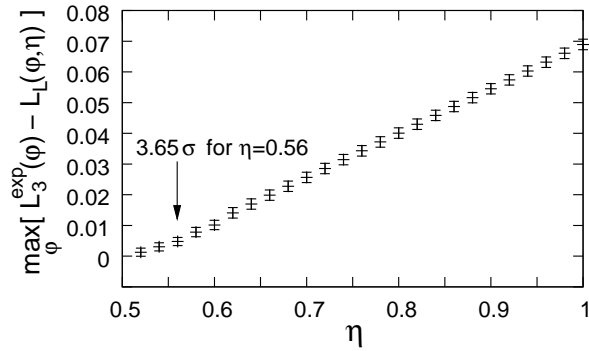
FIG. 4: **Experimental test of the generalized Leggett inequality for different degrees of purity** η**.** From the measurement shown in Figure 3, we extract the maximal excess of the experimental $L_3(\varphi)$ with respect to the upper bound (denoted L_L) in inequality (8), for various degrees of purity η. For $\eta < 0.6$, data at $\varphi = -15°$ are considered, while for $\eta > 0.6$, the largest violation are observed for $\varphi = -25°$.

with quantum mechanics. However, given a model with non-trivial marginals, finding an explicit inequality that can be tested experimentally against quantum predictions is another problem; for that, one needs the specific details of the model.

In summary, with the general goal of improving our understanding of quantum correlations, we reformulated Leggett's model. No time-ordering of the events was assumed, and all assumptions were made on the local part of the correlations. We derived new Leggett-type inequalities, simpler and stronger than previously known. The simplest version of our inequalities has been experimentally violated. Finally we investigated more general models *à la Leggett*, for which we only imposed the no-signaling condition. We argued that any such model with biased marginals is incompatible with quantum predictions for the singlet state. This shows that quantum correlations cannot be reconstructed from "simpler" correlations in which the individual properties would be even partially defined. Nature is really such that, in some cases, individual properties are completely lost while global properties are sharply defined.

Our result is in good agreement with the recent work of Colbeck and Renner [13], who have derived general inequalities to falsify such models with non trivial marginals. An example of a non-signaling model that successfully reproduces the singlet correlations can be found in [14]; indeed this model has unbiased marginals. It is also worth mentioning the nonlocal model of Toner and Bacon [15], which reproduces analytically the singlet correlations with one bit of communication. In this model, the probability distributions P_λ have non vanishing marginals; however, the P_λ's are signaling. The remarkable property of the Toner-Bacon model is that the communication is cleverly hidden, such that the final probability distribution P is non-signaling.

This work is part of the general research program that looks for nonlocal models compatible or incompatible with quantum predictions. The goal is to find out what is essential in quantum correlations. Here we found that in order to simulate or to decompose the singlet correlations, one can't use non-signaling correlations P_λ with non-trivial marginals. This nicely complements the result of [16], that the correlations corresponding to very partially entangled states, hence large marginals, can't be simulated by a single PR-box, which has trivial marginals.

METHODS

A. Simple derivation of inequalities that test Leggett's model

1. Convenient notations

As mentioned, in this paper we focus on the case of binary outcomes $\alpha, \beta = \pm 1$. In this case, the correlations can conveniently be written as

$$P_\lambda(\alpha, \beta | \vec{a}, \vec{b}) = \tag{10}$$
$$\frac{1}{4}\left(1 + \alpha \, M_\lambda^A(\vec{a}, \vec{b}) + \beta \, M_\lambda^B(\vec{a}, \vec{b}) + \alpha\beta \, C_\lambda(\vec{a}, \vec{b})\right).$$

This expression allows one to clearly distinguish the marginals $M_\lambda^A(\vec{a}, \vec{b}) = \sum_{\alpha,\beta} \alpha P_\lambda(\alpha, \beta | \vec{a}, \vec{b})$ on Alice's side and $M_\lambda^B(\vec{a}, \vec{b}) = \sum_{\alpha,\beta} \beta P_\lambda(\alpha, \beta | \vec{a}, \vec{b})$ on Bob's, and the correlation coefficient $C_\lambda(\vec{a}, \vec{b}) = \sum_{\alpha,\beta} \alpha\beta P_\lambda(\alpha, \beta | \vec{a}, \vec{b})$. Throughout the Methods section, we shall use these notations; in the main text, we have used more standard and simplified notations, the correspondence being $\langle\alpha\rangle_\lambda = M_\lambda^A(\vec{a}, \vec{b})$, $\langle\beta\rangle_\lambda = M_\lambda^B(\vec{a}, \vec{b})$. The no-signaling condition is $M_\lambda^A(\vec{a}, \vec{b}) = M_\lambda^A(\vec{a})$ and $M_\lambda^B(\vec{a}, \vec{b}) = M_\lambda^B(\vec{b})$.

In order for the decomposition (1) to be a valid mixture of correlations, all distributions P_λ should be non-negative. As we said, this constraint is enough to derive Leggett-type inequalities. From eq. (10), one can see that the non-negativity implies the general constraints:

$$|M_\lambda^A(\vec{a}) \pm M_\lambda^B(\vec{b})| \le 1 \pm C_\lambda(\vec{a}, \vec{b}) \, . \tag{11}$$

Constraints on the marginals M_λ^A or M_λ^B thus imply constraints on the correlation coefficients C_λ, and *vice versa*.

Let's now consider one measurement setting $\vec{a}$ for Alice and two measurement settings $\vec{b}, \vec{b}'$ for Bob, and let's combine the previous inequalities (11) that we get for $(\vec{a}, \vec{b})$ and $(\vec{a}, \vec{b}')$. Using the triangle inequality, one gets

$$|C_\lambda(\vec{a}, \vec{b}) \pm C_\lambda(\vec{a}, \vec{b}')| \le 2 - |M_\lambda^B(\vec{b}) \mp M_\lambda^B(\vec{b}')| \, . \tag{12}$$

These constraints must hold for all probability distributions P_λ. After integration over the λ's, one

gets, for the averaged correlation coefficients $C(\vec{a}, \vec{b}) = \int d\lambda \rho(\lambda) C_\lambda(\vec{a}, \vec{b})$

$$|C(\vec{a}, \vec{b}) \pm C(\vec{a}, \vec{b}')| \le 2 - \int d\lambda \rho(\lambda) \; |M_\lambda^B(\vec{b}) \mp M_\lambda^B(\vec{b}')| \; . \tag{13}$$

This inequality is general for all models with "local marginals", i.e. that fulfil the no-signaling condition.

2. Derivation of a simple Leggett-type inequality

Now we derive an inequality satisfied by Leggett's specific model, that can be experimentally tested. Inequality (13) implies, for the particular form of eq. (4) for Bob's marginals:

$$|C(\vec{a}, \vec{b}) + C(\vec{a}, \vec{b}')| \le 2 - \int d\lambda \rho(\lambda) \; |\vec{v} \cdot (\vec{b} - \vec{b}')| \; . \tag{14}$$

Let's consider three triplets of settings $(\vec{a}_i, \vec{b}_i, \vec{b}_i')$, with the same angle φ between all pairs $(\vec{b}_i, \vec{b}_i')$, and such that $\vec{b}_i - \vec{b}_i' = 2\sin\frac{\varphi}{2}\vec{e}_i$, where $\{\vec{e}_1, \vec{e}_2, \vec{e}_3\}$ form an orthogonal basis (see Figure 1). After combining the three corresponding inequalities (14), using the fact that $\sum_{i=1}^3 |\vec{v} \cdot \vec{e}_i| \ge 1$ and the normalization $\int d\lambda \rho(\lambda) = 1$, we finally get the Leggett-type inequality (5).

For a pure singlet state, inequality (5) is violated when $|\varphi| < 4\arctan\frac{1}{3} \simeq 73.7°$, and the maximal violation is obtained for $|\varphi| = 2\arctan\frac{1}{3} \simeq 36.9°$. In the case of imperfect interference visibility V $(\tilde{L}_{\Psi^-}(\varphi) = 2V|\cos\frac{\varphi}{2}|)$, a violation can still be observed as long as $V > V_{th}^{(3)} = \sqrt{1 - (\frac{1}{3})^2} = \frac{2\sqrt{2}}{3} \simeq 94.3\%$.

Note that other Leggett-type inequalities can be derived, as we mention in Supplementary Information III.

B. Any non-signaling model must have vanishing marginals

We prove here that all marginals in a non-signaling model must necessarily satisfy the constraints (17) and (18) below; and we argue that this in turn implies the claim made in the main text, namely: all the marginals must vanish (apart perhaps on a zero-measure subset of λ's).

In order for a general non-signaling model to reproduce all quantum correlations $C_{\Psi^-}(\vec{a}, \vec{b}) = -\vec{a} \cdot \vec{b}$ of the singlet state, one must have, according to equation (13), for all $\vec{a}, \vec{b}, \vec{b}'$ on the Poincaré sphere:

$$\int d\lambda \rho(\lambda) \; |M_\lambda^B(\vec{b}) \pm M_\lambda^B(\vec{b}')| \le 2 - |\vec{a} \cdot (\vec{b} \mp \vec{b}')| \tag{15}$$

and therefore, for all $\vec{b}, \vec{b}'$:

$$\int d\lambda \rho(\lambda) \; |M_\lambda^B(\vec{b}) \pm M_\lambda^B(\vec{b}')| \le 2 - ||\vec{b} \mp \vec{b}'|| \tag{16}$$

where $|| \cdot ||$ is the euclidian norm.

In the case where $\vec{b}' = -\vec{b}$, the first constraint of eq. (16) implies that for all $\vec{b}$

$$\int d\lambda \rho(\lambda) \; |M_\lambda^B(\vec{b}) + M_\lambda^B(-\vec{b})| = 0 \; . \tag{17}$$

In the case $\vec{b}' \to \vec{b}$, the two vectors being normalized, we have $2 - ||\vec{b} + \vec{b}'|| = 2 - \sqrt{4 - ||\vec{b} - \vec{b}'||^2} \sim \frac{||\vec{b} - \vec{b}'||^2}{4}$ and therefore the second constraint of (16) implies that for all $\vec{b}$

$$\lim_{\vec{b}' \to \vec{b}} \int d\lambda \rho(\lambda) \; \frac{|M_\lambda^B(\vec{b}) - M_\lambda^B(\vec{b}')|}{||\vec{b} - \vec{b}'||} = 0 \; . \tag{18}$$

Let us now prove, in the case of discrete λ's ($\lambda \in \{\lambda_i\}$), that (17) and (18) in turn necessarily imply that the marginals $M_\lambda^B(\vec{b})$ must vanish. In this case, the integral $\int d\lambda \rho(\lambda)$ should be changed to a discrete sum $\sum_i p_{\lambda_i}$.

Constraint (17) implies indeed that for all λ_i (such that $p_{\lambda_i} > 0$) and for all $\vec{b}$, $M_{\lambda_i}^B(-\vec{b}) = -M_{\lambda_i}^B(\vec{b})$, i.e. $M_{\lambda_i}^B$ must be an odd function of $\vec{b}$. This is a very natural property that one would impose to such a model; in particular, Leggett's model has indeed odd marginals.

In addition, constraint (18) implies that for all λ_i and for all $\vec{b}$, $\lim_{\vec{b}' \to \vec{b}} \frac{|M_{\lambda_i}^B(\vec{b}) - M_{\lambda_i}^B(\vec{b}')|}{||\vec{b} - \vec{b}'||} = 0$, i.e. that all $M_{\lambda_i}^B$ are differentiable, and their derivative is 0 for all $\vec{b}$: therefore they are constant. Since they have to be odd functions, then necessarily they are equal to zero.

In conclusion, for discrete λ's, Bob's marginals $M_{\lambda_i}^B(\vec{b})$ must all vanish; of course, the same reasoning holds for Alice's marginals $M_{\lambda_i}^A(\vec{a})$. This result should also be valid for any distribution $\rho(\lambda)$, at least those physically reasonable (e.g. piecewise continuous): we conjecture that for any reasonable model to reproduce the quantum correlations of the singlet state, necessarily the marginals must vanish, in the sense that

$$\forall \vec{a}, \quad \int d\lambda \rho(\lambda) |M_\lambda^A(\vec{a})| = 0 \; , \tag{19}$$

$$\forall \vec{b}, \quad \int d\lambda \rho(\lambda) |M_\lambda^B(\vec{b})| = 0 \; , \tag{20}$$

i.e for all $\vec{a}, \vec{b}$, $|M_\lambda^A(\vec{a})| = |M_\lambda^B(\vec{b})| = 0$ for "almost all" λ (except for a zero-measure subset of λ's, that could possibly depend on $\vec{a}, \vec{b}$).

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Acknowledgements

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SUPPLEMENTARY INFORMATION

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SUPPLEMENTARY INFORMATION I: TECHNICAL ASPECTS OF THE EXPERIMENT

A. Pair source and detection

The polarization-entangled photon pair source in our experimental setup (Figure 2 of the article) is based on a non-collinear type-II parametric down conversion process in a 2 mm thick properly cut Barium-beta-borate crystal, where photon pairs with almost degenerate wavelengths around 702 nm are collected [1]. As a pump source (PS), we used an Argon ion laser with a center wavelength of 351 nm and a power of 38 mW. Longitudinal and partial transverse walk-off compensation was established in the usual way using a half wave plate and two 1 mm thick BBO crystals (CC) of the same cut as the conversion crystal [2].

For the polarization measurements we used zero order quarter wave plates ($\lambda/4$) and polymer-film based polarization filters (PF) with a small wedge error and an extinction ratio of better than 1:10,000. All four elements could be oriented with motorized rotation stages with a reproducibility of about 0.1°.

Photo events were generated with fiber-coupled passively quenched silicon avalanche photodiodes ($D_{A,B}$). All photo events were recorded, and pairs identified when photo events coincided in a time window of $\tau = 15$ ns.

A pair of interference filters (IF, center wavelength 702 nm, full transmission width 5 nm at half maximum) was used for initial alignment of the pair source, and left in the system to avoid a change in beam orientations.

B. Alignment and setup characterization

As a first step in our alignment procedure, the single mode optical fibers were adjusted for neutral polarization transport with polarization controllers (FPC). Under stable temperature conditions, we maintained an extinction ratio better than 1:1000 over half a day. Then, the exact orientation θ_B of the polarizing filter with respect to the first one was fixed to an accuracy of 0.2° by searching for a minimum of coincidence events, with the quarter wave plates removed. Compensation crystals were adjusted to maximize the visibility of polarization (anti-)correlations

in the $\pm 45°$ basis. At this point, we were reasonably confident that we observe photon pairs in a good approximation of a singlet polarization state. Typical visibilities we reached were $V_{HV} \approx 99\%$ and $V_{\pm 45} \approx 98\%$.

There is no direct evidence in the experimental result for $L_3(\varphi)$ if the orientation of the quarter wave plates with respect to the polarizing filters is accurate. We thus verified the orientation of the first wave plate by symmetrization of the polarization correlations with a fixed polarization filter in Bob's arm under 0° linear polarization, and varying θ_B while observing coincidences. With this procedure, we were able to find the symmetric position within $\Delta\gamma_A \approx 0.2°$. The quarter wave plate in Bob's arm was oriented in a similar way.

To test the inequality (5) of the article, we used the following Poincaré vectors (see Figure 1):

$$\vec{a}_1 = \vec{x}\,, \quad \vec{b}_1/\vec{b}_1' = \left(\cos\frac{\varphi}{2}, \pm\sin\frac{\varphi}{2}, 0\right),$$
$$\vec{a}_2 = \vec{y}\,, \quad \vec{b}_2/\vec{b}_2' = \left(0, \cos\frac{\varphi}{2}, \pm\sin\frac{\varphi}{2}\right),$$
$$\vec{a}_3 = \vec{z}\,, \quad \vec{b}_3/\vec{b}_3' = \left(\pm\sin\frac{\varphi}{2}, 0, \cos\frac{\varphi}{2}\right), \tag{S1}$$

so that

$$\vec{b}_i - \vec{b}_i' = 2\sin\frac{\varphi}{2}\vec{e}_i\,, \text{ with } (\vec{e}_1, \vec{e}_2, \vec{e}_3) = (\vec{y}, \vec{z}, \vec{x}). \tag{S2}$$

The x axis in this notation corresponds to $\pm 45°$ linear, the y axis to circular, and the z axis to horizontal/vertical polarization, the latter coinciding with the natural basis of the parametric down conversion process in the nonlinear optical crystal.

The correlation coefficients $C(\vec{a}, \vec{b})$ for each of the six settings for testing inequality (5) were obtained by recording photo-detection coincidences c during an integration time T for four combinations of orthogonal polarizations by rotating the polarizers by additional 90° accordingly:

$$C(\vec{a}, \vec{b}) = \frac{c(\vec{a}, \vec{b}) + c(-\vec{a}, -\vec{b}) - c(-\vec{a}, \vec{b}) - c(\vec{a}, -\vec{b})}{c(\vec{a}, \vec{b}) + c(-\vec{a}, -\vec{b}) + c(-\vec{a}, \vec{b}) + c(\vec{a}, -\vec{b})}. \tag{S3}$$

This way, artefacts due to imbalanced detector efficiencies were minimized.

In order to ensure the stability of the alignment, we recorded polarization correlations in the HV and $\pm 45°$
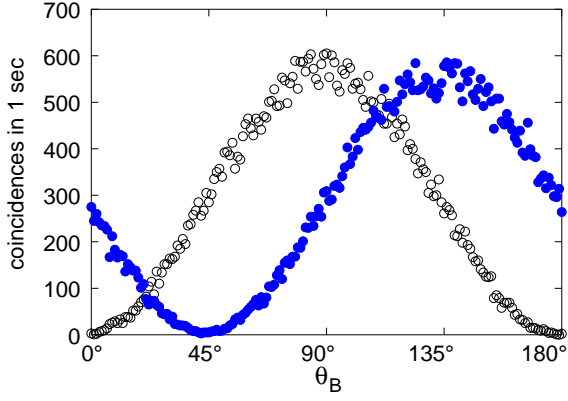
Supplementary Figure 1: Polarization correlations in the HV basis (open circles, $\theta_A = 0°$) and $\pm 45°$ linear basis (filled circles, $\theta_A = 45°$). The corresponding visibilities are $V_{HV} = 98.9 \pm 0.8\%$ and $V_{\pm 45°} = 97.8 \pm 0.8\%$.

linear polarization bases both before and after the acquisition of the correlations for L_3 in the usual way (i.e., $\gamma_A = \theta_A$ fixed, $\gamma_B = \theta_B$ varies). Supplementary Figure 1 shows these polarization correlations directly before the main measurement. We extract visibilities of $V_{HV} = 98.9 \pm 0.8\%$ and $V_{\pm 45°} = 97.8 \pm 0.8\%$, respectively.

The uncertainties in all experimental values of L_3 are obtained assuming Poissonian counting statistics for photodetection events, independent variations for each setting, and the usual error propagation through (S3) and (5).

To track potential drifts of the apparatus, the set shown in Figure 3 of the article was recorded in two interleaving series with a spacing of $5°$ for φ each. The absence of a modulation of $L_3(\varphi)$ with a $5°$ period in the presented graph indicates that the residual asymmetry can not be attributed to drifts of our setup, but to a residual misalignment of the quarter wave plates with respect to each other. From a simulation with a noisy singlet state, the residual asymmetry seems to be compatible with a misalignment on the order of $0.2°$.

The polarization correlations after these measurements revealed values of $V_{HV} = 99.3 \pm 0.8\%$ and $V_{\pm 45°} = 97.8 \pm 0.8\%$, indicating again that the alignment of source, optical fibers and measurement system has not drifted significantly over the measurement time of about $9\,\text{h}$.

SUPPLEMENTARY INFORMATION II: OTHER LEGGETT-TYPE INEQUALITIES

Inequality (5) in the article is certainly the simplest one that allows to test Leggett's model versus quantum mechanics. It involves only 3 settings for Alice and 6 for Bob. Note that on Alice's side, two settings could actually be chosen to be the same: in our case, one could for instance rotate the triplet $(\vec{a}_1, \vec{b}_1, \vec{b}_1')$ around the y axis, so that $\vec{a}_1 = \vec{a}_3 = \vec{z}$. Thus, 2×6 settings are actually

enough [7]. In addition, inequality (5) is stronger than the previously derived inequalities [3, 4, 5, 6]. Consider for instance the simplest inequalities of [5, 6]: in our formalism, they would be obtained by (i) choosing four triplets, in two orthogonal planes, and (ii) setting $\vec{a}_i = \vec{b}_i$. Now, the choice of the triplets was not optimal, nor was the choice (ii), as the geometry of the problem clearly shows.

Following our reasoning, one can derive even more robust inequalities by increasing the number N of (judiciously chosen) triplets $(\vec{a}_i, \vec{b}_i, \vec{b}_i')$, thus defining N directions $\vec{e}_i$ for $\vec{b}_i - \vec{b}_i'$. If $\frac{1}{N} \sum_{i=1}^{N} |\vec{v} \cdot \vec{e}_i| \geq \xi_N > 0$ holds for all $\vec{v}$, then one gets the Leggett-type inequality

$$L_N(\varphi) \leq 2 - 2\,\xi_N\,|\sin \frac{\varphi}{2}|\,, \qquad (S4)$$

with $L_N(\varphi) = \frac{1}{N} \sum_{i=1}^{N} |C(\vec{a}_i, \vec{b}_i) + C(\vec{a}_i, \vec{b}_i')|$. This inequality is violated by the singlet state, for which $L_N(\varphi)$ is still equal to $L_{\Psi^-}(\varphi) = 2|\cos \frac{\varphi}{2}|$. The threshold visibility is now $V_{th}^{(N)} = \sqrt{1 - \xi_N^2}$. The next simple case after the one we studied is $N = 4$: in this case the four directions $\vec{e}_i$ should point to the four vertices of the regular tetrahedron. One can then show that $\frac{1}{4} \sum_{i=1}^{4} |\vec{v} \cdot \vec{e}_i| \geq \frac{1}{\sqrt{6}}$ (i.e. $\xi_4 = \frac{1}{\sqrt{6}}$), and one gets an inequality that tolerates a visibility of $V_{th}^{(4)} = \sqrt{\frac{5}{6}} \simeq 91.3\%$. In the infinite (and experimentally non-testable) limit of all directions $\vec{e}$ scanned by the vectors $\vec{b}_i - \vec{b}_i'$, we would have $\xi_N \to \frac{1}{2}$, and $V_{th}^{(N \to \infty)} = \frac{\sqrt{3}}{2} \simeq 86.6\%$.

SUPPLEMENTARY INFORMATION III: A STRAIGHTFORWARD GENERALIZATION OF LEGGETT'S MODEL

Here we come back to the generalization of Leggett's model, in which we impose that the marginals should have the following form:

$$\begin{aligned} \langle \alpha \rangle_\lambda &= \eta\,\vec{u} \cdot \vec{a}\,, \\ \langle \beta \rangle_\lambda &= \eta\,\vec{v} \cdot \vec{b}\,, \end{aligned} \quad \text{with } 0 < \eta \leq 1\,. \qquad (S5)$$

This generalized model satisfies inequalities similar to (S4):

$$L_N(\varphi) \leq 2 - 2\,\eta\,\xi_N\,|\sin \frac{\varphi}{2}|\,. \qquad (S6)$$

For any value of $\eta > 0$, these inequalities are violated by the quantum mechanical prediction $L_{\Psi^-}(\varphi)$, for a small enough angle φ. Thus, such generalizations of Leggett's model also fail to reproduce quantum mechanical predictions, and can also be tested experimentally.

The required visibility to violate inequalities (S6) depends on η: $V_{th}^{(N)} = \sqrt{1 - (\eta\,\xi_N)^2}$. In the experimental run shown in Figure 3, we had $N = 3$, $\xi_3 = \frac{1}{3}$, and an average visibility of $98.4 \pm 0.4\%$. Therefore our data allows us to falsify these generalized models for all

$\eta > 0.528 \pm 0.07$. The statistical significance of the experimental excess of L_3 with respect to the generalized Leggett bound increases roughly linearly with η, from $3.65\,\sigma$ for $\eta = 0.65$ up to $40.6\,\sigma$ of the Leggett model bound (5) for $\eta = 1$.

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[7] Of course, we do not claim that this number of settings (2×6) is minimal. We have found other inequalities, some with terms of the form $|C(\vec{a}, \vec{b}) - C(\vec{a}, \vec{b}')|$, that use fewer settings but which are less robust.